Influence of Blocking Interlayer in Blue Organic Light-Emitting Diodes with Different Thicknesses of Emitting Layer and Interlayer

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A series of blue organic light-emitting diodes with electron or hole blocking interlayer are fabricated. Different structures of blocking interlayers can influence the position of the recombination zone due to the fact that they confine carriers in different ways. We find that the double hole blocking interlayer structure can balance carrier injection more effectively. Its power and current efficiency are more stable and the current efficiency value is 30.2 cd/A at 1000 cd/m^2. Decreasing the thicknesses of the emitting layer and interlayers is in favor of the power efficiency. The performance of the device is also affected by changing the interlayer position when we use hole and electron blocking material as interlayers simultaneously.

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Nowadays, organic light-emitting diodes (OLEDs) have attracted much attention for their high electroluminescence efficiency, considerable for next-generation panel display and lighting sources, with the advantages of good flexibility and short response time.[1−4] The power efficiency of white OLEDs has recently reached 102 lm/W at 3000 cd/m^2.[5] It is above the efficiency of a general fluorescent tube. Usually, high efficient OLEDs are fabricated with phosphorescent materials. Phosphorescent OLEDs (PHOLEDs) can harvest both singlet and triplet excitons. By using phosphorescent materials, the internal quantum efficiencies close to 100% can be reached.[6−9] However, when the brightness or current density becomes higher, the electroluminescence efficiency of devices will decrease obviously. This is called efficiency roll-off.[10,11] The efficient and stable PHOLEDs rely on the structure which can ensure that the holes and electrons are balanced injected into the emitting-layer (EML). Some structures can effectively improve the device performance, such as mixed-host[12,13] and inset interlayer.[14−16] The interlayer structure usually uses the bipolar interlayer. For more balanced injecting charge into EML, the polar interlayer is needed.

In the present work, we fabricate a series of blue PHOLEDs. These devices use di-[4-(N,N-ditolyl-amino)-phenyl]cyclohexane (TAPC) or 1,3,5-tri[(3-pyridyl)-phen-3-yl]benzene (TmPyPB) as the interlayer in different positions. These two kinds of interlayers have different characteristics to block holes or electrons. All the devices have two interlayers. Then, the devices with different thicknesses are fabricated to discuss the thickness influence. In the end, we discuss the influence of interlayer positions in two devices which use the same materials and thickness.

All the devices are fabricated by vacuum deposition under a base pressure lower than 5×10^{-5} Pa without breaking the vacuum. The substrate is a glass sheet which is coated with indium tin-oxide (ITO). The sheet resistance is 15 Ω/sq. Before deposition, the substrates are cleaned by ultrasonic bath for 15 min in each of the following: detergent, deionized water, propanol. All the organic materials are deposited on the substrates at a rate of 2 Å/s, and the metal cathodes are deposited at a rate of 2 Å/s. The deposition rate and film thickness are monitored by quartz crystal oscillators. The electroluminescence (EL) spectra and luminance characteristic are measured by a PR655 spectrometer. The current density J and voltage V are obtained by using the source and measured by a Keithley 2400 digital power.

TAPC is chosen as the electronic-blocking interlayer due to the high lowest occupied molecular orbital (LUMO) level of 2.0 eV. The hole-blocking interlayer is TmPyPB due to the deep highest occupied molecular orbital (HOMO) level of 6.68 eV. All devices use iridium(Ⅲ)bis(4,6-difluorophenyl-pyridinato-N,C2)picolinate (FIrpic) as blue dopant and 1,3-di(9H-carbazol-9-yl)benzene (mCP) as host material. The basic device structure is: ITO (100 nm)/Dipyrazino[2,3-f: 2',3'-h]quinoxaline-2,3,6,7,10,11-hexacarbonitrile (HAT-CN) (10 nm)/TAPC (20 nm)/4,4',4"-Tris(carbazol-9-yl)triphenylamine (TCTA) (20 nm)/EML1/interlayer 1/EML2/interlayer 2/EML3/TmPyPB (20 nm)/4,7-Diphenyl-1,10-phenanthroline (Bphen) (20 nm)/Al (100 nm). For devices A1, A2 and A3, the structure of every EML is mCP:FIrpic (8 nm, 10%) and both the interlayer thicknesses are 3 nm. For device A1, interlayers 1 and 2 both are TAPC; for device A2, interlayers 1 and 2 both are TmPyPB; for device A3, interlayer 1 is TAPC and interlayer 2 is TmPyPB.

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Another series of devices B1, B2 and B3 only change the thicknesses of EMLs and interlayers based on devices A1, A2 and A3. The thicknesses of EMLs and interlayers in devices B1, B2 and B3 are half the thicknesses in devices A1, A2 and A3, respectively. In the end, device C1 is fabricated, which is the same as device B3 except the interlayer position.

Table 1. Electroluminescence characteristics of devices tested with different structures.

Devices	Max PE[a] (lm/W)	Max CE[b] (cd/A)	CE at 10^2 cd/m^2 (cd/A)	CE at 10^3 cd/m^2 (cd/A)	Luminance at 8 V (cd/m^2)
A1	29.6	34.6	33.2	26.4	2298
A2	27.3	33.4	33.0	30.2	4772
A3	29.5	33.3	31.2	27.3	3233
B1	28.8	32.1	31.2	23.6	3234
B2	29.2	34.0	33.9	30.3	4160
B3	30.2	33.6	31.1	23.6	3276
C1	29.6	34.2	33.1	26.4	3702

[a]PE is the power efficiency. [b]CE is the current efficiency.

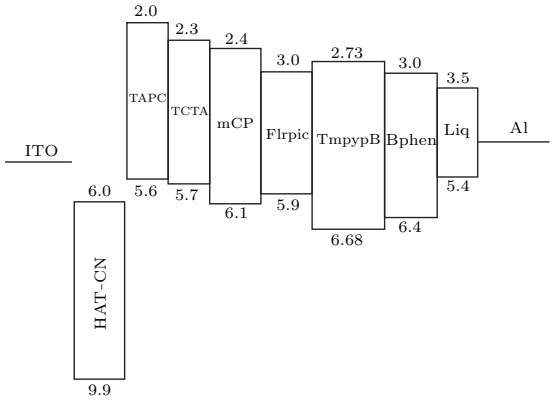

Fig. 1. Energy level diagrams of all materials used in the devices.

Figure 1 shows the energy level diagrams of all materials used in the devices. Table 1 lists the electroluminescence characteristic values of the devices tested with different structures. The electroluminescence characteristic curves of devices A1, A2 and A3 are shown in Fig. 2. It can be seen that the current density is greatly affected by blocking materials. As shown in Fig. 2(a), device A2 has the highest current density. This is due to the fact that mCP is a hole transporting host material, and the hole and electron mobilities are 1.2×10^{-4} cm^2/Vs and 4×10^{-5} cm^2/Vs, respectively.[17] TAPC is an electron blocking material, which makes the electron transport more difficult than TmPyPB. Thus the device which contains the TAPC interlayer obtains a lower current density. However, the current density of device A1 is higher than that of device A2 when the voltage is lower than 3.75 V. This is due to the fact that both hole and electron injections are difficult at low voltage, the effect of transportation is more important than that of blocking, thus it is easier to obtain a higher current density for the TAPC interlayer due to its better hole transportation capability. In addition, the current density of device A1 is higher than that of device A3 at low voltage, and it further proves that the blocking effect of TAPC interlayer is weak at low voltage and becomes stronger with voltage increasing gradually. Although more excitons are produced in device A2 with a higher current density, triplet–triplet annihilation is suppressed due to the excitons scattered in three EMLs in device A3, thus the maximum luminance of device A3 is higher than that of device A2.

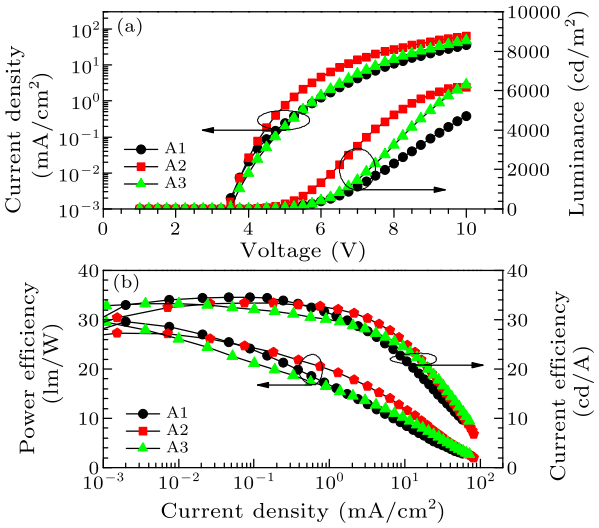

Fig. 2. (a) Current density and luminance versus voltage curves of devices A1, A2 and A3. (b) Power efficiency and current efficiency versus current density curves of devices A1, A2 and A3.

Due to the excitons scattering, the efficiency roll-off is moderated with the current density increasing, thus device A3 is more stable. However two kinds of interlayers in device A3 make the carrier too dispersive, the current efficiency is lower than that in devices A1 and A2. Since the carriers in device A1 are almost accumulated around the side of EML, the highest current efficiency is obtained, while the efficiency roll-off becomes severe with the increasing concentration of excitons. The mCP is the host material which conduces to hole transportation, TmPyPB is the blocking material which conduces to electron transportation,

and device A2 makes the two kinds of carriers more balanced in EML, thus a higher and better stable current efficiency is obtained as shown in Fig. 3.

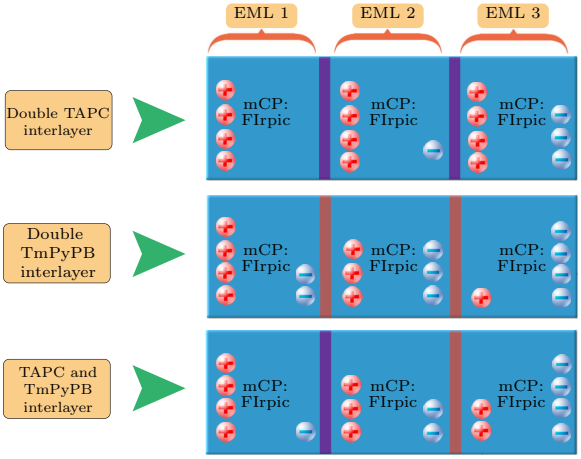

Fig. 3. The function of the blocking interlayer.

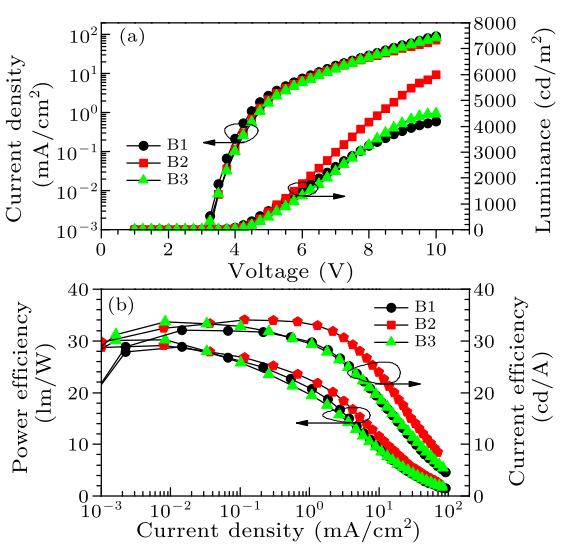

Fig. 4. (a) Current density and luminance versus voltage curves of devices B1, B2 and B3. (b) Power efficiency and current efficiency versus current density curves of devices B1, B2 and B3.

In addition, there is a relationship between the current efficiency and the main emission zone. In device A1, it is difficult for electrons to accumulate in EML1, thus the main emission zone locates in EML2 and EML3 when the current efficiency is the maximal value. In device A2, holes and electrons can uniformly distribute in three EMLs, thus the main emission zone also locates in three EMLs and has a slight change with the increasing current density. This is another reason why the current efficiency of device A2 is stable. Although the current efficiency of device A3 is lower than those of devices A1 and A2, the efficiency roll-off is more moderate due to the fact that the main emission zone of device A3 extends from EML2 to EML1 and EML3 when the current density exceeds 2 mA/cm^2.

The electroluminescence characteristics of devices B1, B2 and B3 are shown in Fig. 4. The curves of current density versus voltage of devices B1, B2 and B3 are similar. With reducing the thickness, the influence of blocking is weakened and the balance of injection is broken, luminances of devices B1, B2 and B3 are lower than those of devices A1, A2 and A3.

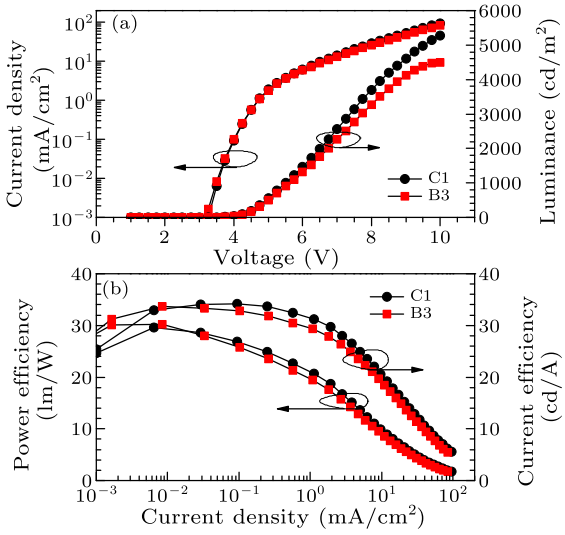

Fig. 5. Current density and luminance versus voltage curves of devices C1 and B3. (b) Power efficiency and current efficiency versus current density curves of devices C1 and B3.

Carrier balance is more important than carrier quantity in devices A1 and B1, thus the power and current efficiencies of device B1 are lower than A1 due to the unbalanced carrier injection. This is due to the fact that the thin EML and interlayer can obtain a higher current density easily, the power and current efficiencies of devices B2 and B3 are both higher than those of devices A2 and A3. Device B3 obtains the maximum current efficiency at low current density, while it decreases rapidly as shown in Fig. 4(b). This is due to the fact that device B3 makes the carriers accumulate in the middle EML easily, the effective recombination results in a higher current efficiency, but the efficiency roll-off becomes severe due to the accumulation of excessive excitons in thinner EML. Device B2 obtains a lower current efficiency than B3 at low current density, while the current efficiency becomes higher and more stable with the increasing current density. The reason is that the combination effect will be enhanced with increasing current density and carrier injection in every EML.

Finally, we fabricate device C1 which exchanges positions of interlayers 1 and 2 based on device B3. The electroluminescence performances of devices C1 and B3 are shown in Fig. 5. The current densities of two devices are similar due to the same EML and interlayer material. At low current density, device C1

obtains a low current efficiency since a small quantity of carrier is injected and accumulates around two sides of EML, effective recombination is decreased. However, the greater the carriers and recombination will become with increasing the current density. At high current density, its current efficiency is higher than that of device B3 since it can more effectively disperse excitons, thus device C1 also obtains a higher and more stable current efficiency.

By experiments, we find that using the different blocking materials as the interlayer can effectively promote the carriers balance in EML. Interlayers can influence the recombination zone position in different ways to confine the carriers, they make the excitons distribute in different EMLs and relax the efficiency roll-off. The device with double TAPC interlayers obtains the highest current efficiency, while it has a severe efficiency roll-off. The device with double TmPyPB interlayers obtains a high and stable current efficiency. The device with TAPC and TmPyPB interlayers has a slow efficiency roll-off at high current density.

In addition, a series of devices with thin EML and interlayer have also been fabricated. The influence of thickness on blocking effect and current density is discussed. The device with double TmPyPB interlayers obtains high and stable power efficiency due to the balanced carrier injection. Finally, the influence of interlayer position is discussed under the condition with the same materials and thickness. In the thinner device, the current efficiency is higher and more stable when the hole blocking material interlayer is near the hole injection side and the electron blocking material interlayer is near the electron injection side.

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